

Fano q reversal in topological insulator Bi_2Se_3

S.V. Dordevic

E-mail: dsasa@uakron.edu

Department of Physics, The University of Akron, Akron, Ohio 44325 USA

G.M. Foster

Department of Physics, The University of Akron, Akron, Ohio 44325 USA

M.S. Wolf

Department of Physics, The University of Akron, Akron, Ohio 44325 USA

N. Stojilovic

Department of Physics and Astronomy, University of Wisconsin Oshkosh, Oshkosh, Wisconsin 54901 USA

Hechang Lei*

Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973 USA

C. Petrovic

Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973 USA

Z. Chen

National High Magnetic Field Laboratory, Tallahassee, Florida 32310 USA

Z.Q. Li

National High Magnetic Field Laboratory, Tallahassee, Florida 32310 USA

L.C. Tung

Department of Physics and Astronomy, University of North Dakota, Grand Forks, North Dakota 58202 USA

National High Magnetic Field Laboratory, Tallahassee, Florida 32310 USA

Abstract. We studied magneto-optical response of a canonical topological insulator Bi_2Se_3 with the goal of addressing a controversial issue of electron-phonon coupling. Magnetic-field induced modifications of reflectance are very pronounced in the infrared part of the spectrum, indicating strong electron-phonon coupling. This coupling causes

an asymmetric line-shape of the 60 cm^{-1} phonon mode, and is analyzed within the Fano formalism. The analysis reveals that the Fano asymmetry parameter (q) changes sign when the cyclotron resonance is degenerate with the phonon mode. To the best of our knowledge this is the first example of magnetic field driven q -reversal.

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Coupling of optical phonons to charge carriers in topological insulators is a topic of considerable current interest [1]. The topic is important from the fundamental point, as well as for potential practical applications of topological insulators. The problem has been studied with a variety of experimental techniques, including optical spectroscopy [2, 3, 4], Angle Resolved Photoemission Spectroscopy [5, 6, 7], time-domain THz spectroscopy [8] and helium scattering [9]. The importance of phonons has also been studied theoretically [10]. Majority of the measurements have been done on a canonical 3D topological insulator Bi_2Se_3 , but the results have been contradictory. Namely, the reported values of the electron-phonon coupling constant λ vary in a broad range, from exceptionally weak [6] to moderately strong [8].

In this work we use magneto-optical spectroscopy to address this controversial issue from another angle. Our magneto-optical results reveal that charge carriers are indeed coupled to 60 cm^{-1} optical phonon, and the coupling is causing an asymmetric line shape of the phonon. Moreover, we find that as the magnetic field increases, the lineshape asymmetry parameter (q) changes sign, from negative to positive. This effect is known from other branches of physics, and is referred to as the q -reversal. We show that the origin of q -reversal in Bi_2Se_3 is the coupling of the phonon to a cyclotron resonance, whose frequency increases linearly with magnetic field.

Single crystals of Bi_2Se_3 were grown at Brookhaven National Laboratory [11, 12]. The samples were characterized with X-ray diffraction using a Rigaku Miniflex X-ray machine. The analysis showed that samples were single phase, and with lattice parameters consistent with the published values [13]. Samples had a typical thickness of several millimeters and naturally flat surfaces, with surface area of about $4 \text{ mm} \times 4 \text{ mm}$. Surfaces were cleaved before each spectroscopic measurement.

Far-infrared magneto-reflectance ratios $R(\omega, B)/R(\omega, 0 \text{ T})$ of Bi_2Se_3 were collected at the National High Magnetic Field Laboratory in magnetic fields as high as 18 T. Magneto-optical spectra of Bi_2Se_3 have previously been reported only in magnetic field up to 8 T [2, 3, 14, 15, 8] and only recently in higher magnetic field [16]. The reflectance ratios $R(\omega, B)/R(\omega, 0 \text{ T})$ were supplemented with the zero field data to obtain the absolute values of reflectance in magnetic field $R(\omega, B)$, as the product of the ratios with the absolute value of reflectance in zero field [17]. Zero magnetic field far-infrared measurements on Bi_2Se_3 were performed at The University of Akron, and have been reported previously [18]. All measurements were performed with unpolarized light, with the electric field vector parallel to the planes.

Figure 1(a) shows the far-infrared reflectance ratios $R(\omega, B)/R(\omega, 0 \text{ T})$ in magnetic field up to 18 T, in steps of 1 T. These ratios provide the most direct evidence for magneto-optical sensitivity of Bi_2Se_3 in the far-infrared part of the spectrum. Large field-induced changes, as high as 75 %, are detected around 220 cm^{-1} , much larger than reported previously [2]. This is another testimony to the excellent sample quality, which when combined with a high magnetic field allowed for the first time [2, 3, 14] the observation of a cyclotron resonance in bulk Bi_2Se_3 samples. The cyclotron resonance in Bi_2Se_3 has so far been observed only in thin films [8, 16].

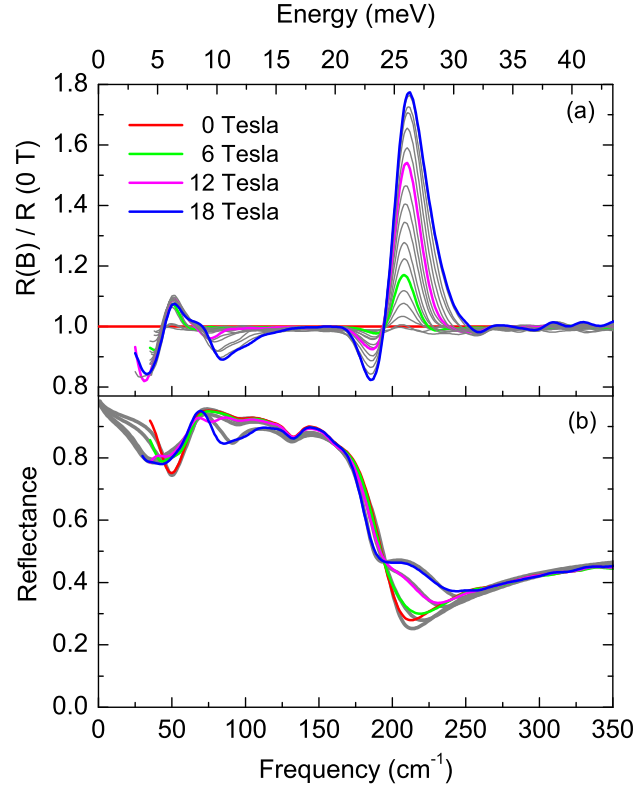


Figure 1. (Color online). (a) Magneto-reflectance ratios $R(\omega, B)/R(\omega, 0 \text{ T})$ for Bi_2Se_3 in fields up to 18 T. Very strong field induced changes are detected around the plasma minimum, as well as around the 60 cm^{-1} phonon mode. (b) The absolute values of reflectance in magnetic field $R(\omega, B)$, obtained from the ratios shown in panel (a) and the absolute values of reflectance in zero field [17, 18]. Gray lines are the best fits obtained from Eqs. 1 and 2.

Figure 1(b) displays the absolute values of reflectance $R(\omega, B)$ obtained from the ratios and the zero-field absolute values. In zero field [18], the most prominent feature in reflectance is the plasma edge located at approximately 180 cm^{-1} . In addition, two optically active phonon modes, observed previously [3, 2, 14], are detected in the form of dips located at approximately 60 cm^{-1} and 130 cm^{-1} . As magnetic field is applied the reflectance is altered dramatically, with the largest changes induced around the plasma minimum. In addition, we also detect relatively large changes in the very far-infrared part of the spectrum, around 60 cm^{-1} phonon mode.

Kramers-Kronig transformation is not directly applicable to magneto-optical spectra [19], so instead we apply the fitting procedures, and from the best fits we generate the optical functions of interest. The most frequently used model is the Drude-Lorentz model, supplemented with the cyclotron resonance [20]. The dielectric function tensor $\varepsilon_{\pm}(\omega) = \varepsilon_{xx} \pm i\varepsilon_{xy}$ is usually represented as:

$$\varepsilon_{\pm}(\omega) = \varepsilon_{\infty} + \sum_i \frac{\omega_{p,i}^2}{\omega_{0,i}^2 - \omega^2 - i\gamma_i\omega \mp \omega_{c,i}\omega} \quad (1)$$

where ε_{∞} is the high-frequency dielectric constant, ω_0 is the central frequency of the

mode, ω_p its oscillator strength and γ its width. ω_c is the cyclotron frequency, which is usually taken as positive for electrons, and negative for holes. Since charge carriers in Bi_2Se_3 have been known to be electrons [3], the cyclotron frequency will be taken as positive, and the cyclotron resonance will show up at positive frequencies in $\varepsilon_-(\omega)$.

To achieve satisfactory fits and to capture the most important features of the data a minimal model consisting of three modes is necessary. These three modes are: 1) a Drude mode, which in magnetic field shifts to finite frequencies and becomes a cyclotron resonance, 2) a phonon mode at 60 cm^{-1} , which has been known to be asymmetric [2, 18], and 3) a phonon mode at 130 cm^{-1} which does not have significant asymmetry and is modeled as a Lorentzian [18]. The cyclotron mode was modeled with Eq. 1, assuming $\omega_0=0$ for all fields. The asymmetric 60 cm^{-1} phonon was modeled as a Fano mode [25, 21]:

$$\varepsilon_F(\omega) = \frac{\omega_{p,F}^2}{\omega_{0,F}^2 - \omega^2 - i\gamma_F\omega} \left(1 + i\frac{\omega_q}{\omega} \right)^2 + \left(\frac{\omega_{p,F}\omega_q}{\omega_{0,F}\omega} \right)^2 \quad (2)$$

where $\omega_{0,F}$, $\omega_{p,F}$ and γ_F are the central frequency, oscillator strength and width of the Fano mode, respectively. ω_q is the Fano frequency, related to the usual Fano asymmetry parameter q as $q=\omega_{0,F}/\omega_q$. As discussed previously [25], large values of $|q|$ (i.e. small values of ω_q) indicate little or no asymmetry, whereas small values of $|q|$ (both positive and negative) indicate large degree of asymmetry. The form of the Fano dielectric function given by Eq. 2 was introduced specifically for infrared spectra [21] and has several advantages over the conventional form [25]. In particular, the special case of no asymmetry (i.e. a Lorentzian lineshape) is achieved for $\omega_q=0$, instead of $q \rightarrow \infty$.

The results of the fits at several selected fields are shown in Fig. 1(b) with gray lines. As can be seen from the plot, the model is capable of capturing all the essential features of the data at all fields. This is significant, as the changes of reflectance induced by magnetic field are very large and dramatic. Namely, as the field increases, the plasma edge is gradually altered and a characteristic "second plasma edge" develops around 220 cm^{-1} , similar to what was observed in Bi [26] and $\text{Bi}_{1-x}\text{Sb}_x$ [27]. In addition, at the lowest frequencies the reflectance is suppressed as the cyclotron resonance enters the measured frequency window. We note that the quality of the fits is similar at different magnetic fields, as exemplified through χ^2 measures, which are all within 10 % [28].

From the best fits of reflectance obtained with Eqs. 1 and 2 we can now generate other optical functions of interests, in particular the circular conductivities $\sigma_{\pm}(\omega) = \omega(\varepsilon_{\pm}(\omega) - 1)/(4\pi i)$. As an example in Fig. 2 we displays the real part of circular conductivity $\sigma_-(\omega)$ at 18 T. Also shown are the individual components of the fits: the cyclotron resonance, the asymmetric 60 cm^{-1} phonon mode and the 130 cm^{-1} phonon.

Overall, the optical conductivity $\sigma_-(\omega)$ is dominated by the phonon mode at 60 cm^{-1} . In zero field there is also a Drude-like mode, but as the magnetic field increases this mode is gradually suppressed [15] and shifts to finite frequencies to become a cyclotron resonance. For fields below approximately 5 T, the resonance is outside of our measured frequency window ($\omega \geq 40 \text{ cm}^{-1}$, see Fig. 1), and only its tail is visible.

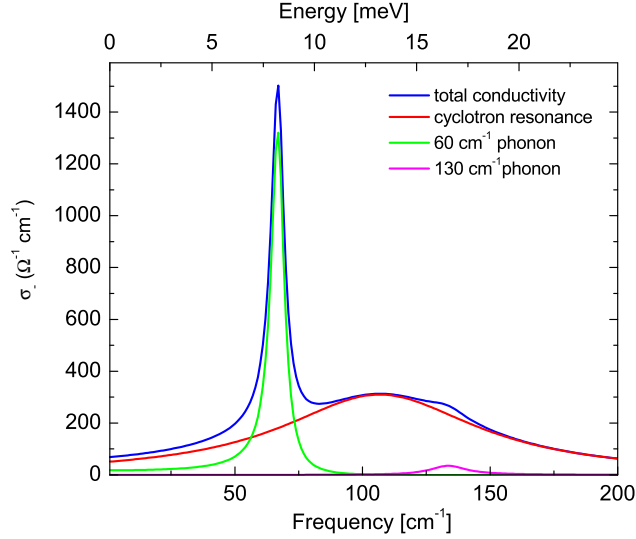


Figure 2. (Color online). Circular optical conductivity $\sigma_{-}(\omega)$ generated from the best fits (Eqs. 1 and 2) at 18 T is shown with blue line. Also shown are the individual components of the fits: the cyclotron resonance (red), the asymmetric 60 cm^{-1} phonon mode (green) and the 130 cm^{-1} phonon (magenta). Note that the cyclotron resonance is several times broader than the phonons.

As field increases, the mode gradually shifts to higher frequencies, its center frequency progressing as a linear function of the field (see Fig. 3(a) below), eventually reaching 110 cm^{-1} (13.6 meV) in 18 T. We point out that the cyclotron resonance is degenerate with the 60 cm^{-1} phonon mode for magnetic field of approximately 10–11 T.

The parameters of the best fits from Eqs. 1 and 2 are shown in Fig. 3. Fig. 3(a) displays the cyclotron resonance frequency ω_c (full squares). The other parameters of the fits are not shown because they are field independent, within the error bars of the fits. On the other hand, the cyclotron frequency ω_c displays a characteristic linear field dependence which extrapolates to zero in zero field. A linear fit (shown with a dashed line) yields $\hbar\omega_c/B = 0.77 \text{ meV/T}$, from which we extract the cyclotron effective mass $m^*/m_e = 0.15$. This value is in excellent agreement with the value (0.14) extracted from a recent magnet-transmission measurement on a thick film [16]. A fraction of this data are shown in Fig. 3(a) with open circles. These values of effective mass are approximately 25 % smaller compared to the value reported for a Bi_2Se_3 thin films [8]. The authors of Ref. [8] used time-domain THz spectroscopy in magnetic fields up to 7 T and obtained the effective mass $m^*/m_e = 0.20$. Their data are shown in Fig. 3(a) with open squares. These recently reported values are in general agreement with earlier reports from optical and quantum oscillations measurements [22, 23, 24].

Overall the agreement between these three data sets is significant, considering that they were obtained using different experimental techniques. Moreover, our measurements were done on a bulk Bi_2Se_3 , whereas the other two were performed on films (of very different thicknesses). We also estimate carrier mobility in our sample $\mu = \omega_c/(B \gamma) = 1800 \text{ cm}^2/\text{Vs}$, which is almost a factor of 2 smaller compared with the

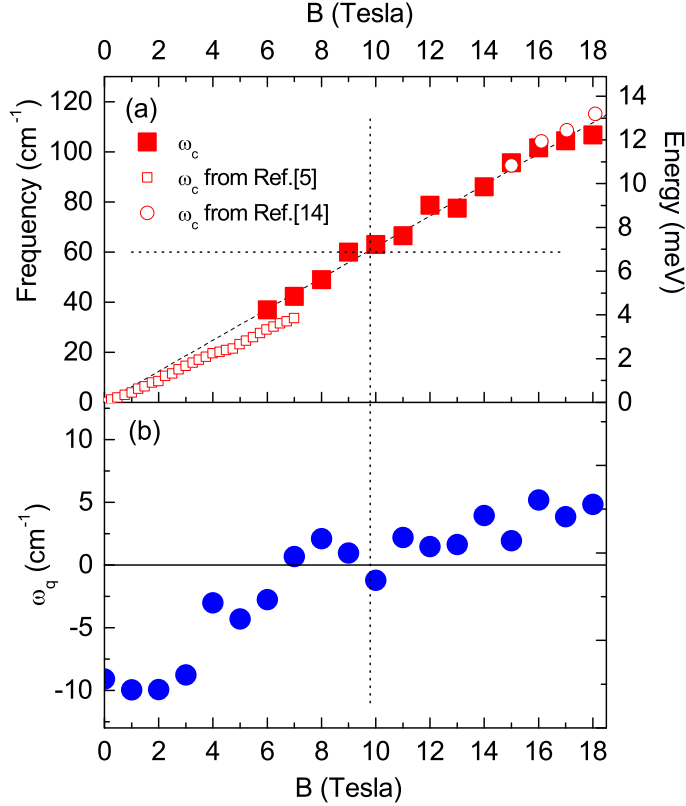


Figure 3. (Color online). Parameters of the best fits of magneto-reflectance from Eqs. 1 and 2. (a) Cyclotron resonance frequency ω_c is shown with full squares, along with the linear fit (dashed line). Also shown is the cyclotron frequency obtained on thin films: open squares from Ref. [8] and open circles from Ref. [16]. The horizontal dotted line represents the frequency of the phonon. (b) Fano frequency ω_q , related to the Fano asymmetry parameter q as $q = \omega_{0,F}/\omega_q$. Note that ω_q changes sign at approximately 10 Tesla (vertical dotted line), the same field at which the cyclotron frequency is degenerate with the phonon mode.

value obtained on the thin film [8]. However, our value is comparable with the values obtained from Hall measurements [3] on bulk Bi_2Se_3 samples with similar carrier density [18], and almost a factor of 2 greater compared with the thick film [16].

Fig. 3(b) shows the Fano frequency ω_q of the 60 cm^{-1} phonon mode and represents the most important finding of the paper. We choose to display ω_q instead of q , because for fields around 10 Tesla, q acquires large values (theoretically it diverges), which renders the fitting unstable. As the plot indicates, ω_q starts as negative [2, 18] for zero and small fields, but around 10 Tesla changes sign and becomes positive. This effect is referred to as the q -reversal. As can be seen from Fig. 3 the asymmetry parameter changes sign when the cyclotron resonance is degenerate with the phonon (phonon frequency is shown with a horizontal dotted line in Fig. 3(a)).

Theory of the asymmetric shape of spectral lines was developed by Fano [25],

and subsequently used in a number of different branches of physics. Examples of the so-called Fano resonance have been found in studies of electron-phonon coupling in superconductors [29], scattering in photonic crystals and plasmonic nanostructures [30], atomic photoemission spectra [31], and many other branches of physics and chemistry. More recently asymmetric Fano lines have been found in the infrared spectra of topological insulator Bi_2Se_3 [2], a few layer graphene [32] and absorption lines of autoionizing helium [33], to name a few.

The degree of lineshape asymmetry is quantified through the Fano parameter q (or alternatively ω_q). In some cases it was observed that, as a results of some external stimulus, the Fano parameter changes sign. This effect is called q -reversal and has been observed for example in atomic physics [34], quantum dots [35], photonics [36] and quantum waveguides [37]. However, Fano q -reversals could not be observed in the previous magneto-optical studies of Bi_2Se_3 [2, 8] due to the limited values of magnetic field ($B \leq 8$ T). The observation of reversal in this study was enabled by large magnetic fields available at the National High Magnetic Field Laboratory. Moreover, we can trace the origin of reversal to the field dependence of the cyclotron resonance (Fig. 3).

In summary, our magneto-optical results on Bi_2Se_3 in magnetic fields up to 18 T reveal that electron-phonon coupling is significant and is causing asymmetric (Fano-like) shape of the 60 cm^{-1} phonon. The asymmetry parameter (q) of the phonon is a function of magnetic field, and around 10 T changes sign, exactly at the field at which the cyclotron resonance is degenerate with the phonon. To the best of our knowledge, this is the first example of Fano q -reversal induced by externally applied magnetic field.

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* Present address: Department of Physics, Renmin University, Beijing 100872, China

References

- [1] Qi X-L, Hughes T L and Zhang S-C 2008 *Physical Review B* **78** 195424
- [2] LaForge A D, Frenzel A, Pursley B C, Lin T, Liu X, Shi J and Basov D N 2010 *Physical Review B* **81** 125120
- [3] Butch N P, Kirshenbaum K, Syers P, Sushkov A B, Jenkins G S, Drew J D and Paglione J 2010 *Physical Review B* **81** 241301(R)
- [4] Reijnders A A, Tian Y, Sandilands L J, Pohl G, Kivlichan I D, Frank Zhao S Y, Jia S, Charles M E, Cava R J, Alidoust N, Xu S, Neupane M, Hasan M Z, Wang X, Cheong S W and Burch K S 2014 *Physical Review B* **89** 075138
- [5] Hatch R C, Bianchi M, Guan D, Bao S, Mi J, Iversen B B, Nilsson L, Hornekaer L, and Hofmann P 2011 *Physical Review B* **83** 241303

- [6] Pan Z-H, Fedorov A V, Gardner D, Lee Y S, Chu S and Valla T 2012 *Physical Review Letters* **108** 187001
- [7] Chen C, Xie Z, Feng Y, Yi H, Liang A, He S, Mou D, He J, Peng Y, Liu X, Liu Y, Zhao L, Liu G, Dong X, Zhang J, Yu L, Wang X, Peng Q, Wang Z, Zhang S, Yang F, Chen C, Xu Z and Zhou X J 2013 *Scientific Reports* **3** 2411
- [8] Wu L, Tse W-K, Brahlek M, Morris C M, Aguilar R V, Koirala N, Oh S and Armitage N P 2015 *Physical Review Letters* **115** 217602
- [9] Zhu X, Santos L, Howard C, Sankar R, Chou F C, Chamon C and El-Batanouny M 2012 *Physical Review Letters* **108** 185501
- [10] Saha K, Legare K and Garate I 2015 *Physical Review Letters* **115** 176405
- [11] Fisk Z and Remeika J P 1989 *Handbook on the Physics and Chemistry of Rare Earths*, edited by Gschneider K A and Eyring J (Amsterdam:Elsevier), Vol. 12
- [12] Canfield P C and Fisk Z 1992 *Philosophical Magazine B* **65** 1117
- [13] Hunter B, *Rietica - A visual Rietveld program*, International Union of Crystallography Commission on Powder Diffraction Newsletter No. 20, (Summer) <http://www.rietica.org> (1998)
- [14] Sushkov A B, Jenkins G S, Schmadel D C, Butch N P, Paglione J and Drew H D 2010 *Physical Review B* **82** 125110
- [15] Aguilar R V, Stier A V, Liu W, Bilbro L S, George D K, Bansal N, Wu L, Cerne J, Markelz A G, Oh S and Armitage N P 2012 *Physical Review Letters* **108** 087403
- [16] Orlita M, Piot B A, Martinez G, Sampath Kumar N K, Faugeras C, Potemski M, Michel C, Hankiewicz E M, Brauner T, Drasar C, Schreyeck S, Grauer S, Brunner K, Gould C, Brune C and Molenkamp L W 2015 *Physical Review Letters* **114** 186401
- [17] Dordevic S V, Komiya S, Ando Y, Wang Y J and Basov D N 2005 *Physical Review B* **71** 054503
- [18] Dordevic S V, Wolf M S, Stojilovic N, Lei H and Petrovic C 2013 *J. Phys.: Condens. Matter* **25** 075501
- [19] Levallois J, Nedoliuk I O, Crassee I and Kuzmenko A B 2015 *Reviews of Scientific Instruments* **86** 033906
- [20] Lax B and Mavroides J G 1967 *Semiconductors and Semimetals*, edited by R.K. Willardson and A.C. Beer, Academic Press, New York and London
- [21] A.B. Kuzmenko A B 2014 *RefFIT manual*
- [22] Groth R und Schnabel P 1964 *Journal of Physics and Chemistry of Solids* **25** 1261
- [23] Tichy L and Borak J 1979 *Physical Review B* **19** 1126
- [24] Kulbachinskii V A, Miura N, Nakagawa H, Arimoto H, Ikaida T, Lostak P and Drasar C 1999 *Physical Review B* **59** 15733
- [25] Fano U 1961 *Physical Review* **124** 1866
- [26] Koncz A, LaForge A D, Li Z, Frenzel A, Pursley B, Lin T, Liu X, Shi J, Dordevic S V and Basov D N 2010 <http://meetings.aps.org/link/BAPS.2010.MAR.H15.4>
- [27] Dordevic S V, Wolf M S, Stojilovic N, Nikolic M V, Vujatovic S S, Nikolic P M and L.C. Tung L C 2012 *Physical Review B* **86** 115119
- [28] The values of χ^2 for several characteristic fields are $\chi^2(0 \text{ T}) = 0.0521348$, $\chi^2(6 \text{ T}) = 0.0473315$, $\chi^2(12 \text{ T}) = 0.048939$ and $\chi^2(18 \text{ T}) = 0.0504891$.
- [29] *Light Scattering in Solids vi*, edited by Cardona M and Guntherodt G 1991 Springer-Verlag Berlin Heidelberg
- [30] Miroshnichenko A E, Flach S and Kivshar Y S, 2010 *Reviews of Modern Physics* **82** 2257
- [31] , Fano U and Rau A R P 1986 *Atomic collisions and spectra* Academic Press
- [32] Li Z, Lui C H, Cappelluti E, Benfatto L, Mak K F, Carr G L, Shan J and Heinz T F 2012 *Physical Review Letters* **108** 156801
- [33] Ott C, Kaldun A, Raith P, Meyer K, Laux M, Evers J, Keitel C H, Greene C H and Pfeifer T 2013 *Science* **340** 716
- [34] Connerade J P, Lane A M and Baig M A 1985 *Journal of Physics B: Atomic, Molecular and Optical Physics* **18** 3507

- [35] Kobayashi K, Aikawa H, Katsumoto S and Iye Y 2002 *Physical Review Letters* **88** 256806
- [36] Driessen E F C, Stolwijk D and de Dood M J A 2007 *Optics Letters* **32** 3137
- [37] Klaiman S, Moiseyev N and Sadeghpour H R 2007 *Physical Review B* **75** 113305